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54 **Antifreeze composition.**

57 An antifreeze composition comprising a glycol and water, containing (A) a phosphoric acid compound, (B) a compound of at least one metal selected from the group consisting of manganese and magnesium, (C) a corrosionproofing agent, and optionally (D) at least one compound selected from the group consisting of (a) an aromatic polybasic acid or a salt thereof, (b) an aliphatic dicarboxylic acid or a salt thereof, (c) a molybdate, (d) a tungstate, (e) a homopolymer derived from an alkylene glycol monoallyl ether or a maleic type monomer, and (f) a copolymer thereof with an other monomer and having a pH value in the range of 6.5 to 9.

## Description

## ANTIFREEZE COMPOSITION

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## BACKGROUND OF THE INVENTION

## Field of the Invention:

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This invention relates to an antifreeze composition. Particularly, it relates to an antifreeze composition composed of a glycol, water, and a corrosion inhibiting agent and used for preventing the cooling water in the liquid-cooling type internal combustion engine from being frozen. More particularly, it relates to an antifreeze composition which, as used in a cooling water for an automobile engine, manifests the effect thereof not merely in preventing the cooling water from being frozen but also in enabling the cooling system of the automobile engine to maintain the function of protecting the automobile engine against rusting and corrosion.

## Description of the Prior Art:

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Heretofore, the cooling liquid for the liquid-cooling type internal combustion engine such as, for example, the automobile engine has heretofore used customarily, for the prevention of the phenomenon of freezing during the cold season, an antifreeze which uses an alcohol for a glycol as a main ingredient and additionally incorporates a varying corrosion-inhibiting agent to acquire an ability to prevent freezing and corrosion at the same time.

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Examples of the alcohol generally used for this purpose include methyl alcohol, ethyl alcohol, and isopropyl alcohol and examples of the glycol generally adopted include ethylene glycol, propylene glycol, hexylene glycol, and glycerol. They are used either singly or in the form of a combination of two or more members. The antifreeze which uses as its main ingredient monoethylene glycol among other compounds cited above finds popular utility in the cooling liquid for the cooling system in the automobile engine.

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In the case of an aqueous monoethylene glycol solution, the effect in preventing the freezing is obtained to  $-15.5^{\circ}\text{C}$  with a concentration of 30% by volume and to  $-37.0^{\circ}\text{C}$  with a concentration of 50% by volume.

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Alcohols or glycols are oxidized on contact with air to produce acidic alcohols or oxides of glycols. The formation of such oxides of alcohols or glycols is all the more accelerated at an elevated temperature in the range of  $50^{\circ}$  to  $100^{\circ}\text{C}$ . These acidic alcohols or glycols promote corrosion of the cooling system in the internal combustion engine, particularly the various metal materials used in the automobile engine. The corrosion of the various metal materials of which the cooling system of the internal combustion engine is made is liable to degrade the thermal conductivity of the cooling system or clog the radiator tube with deposition of the product of corrosion and eventually cause an overheating of the engine.

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The antifreeze which uses an alcohol or a glycol as a main ingredient thereof has to incorporate additionally therein a corrosion-inhibiting agent because the alcohol or glycol is devoid of an ability to prevent the corrosion.

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The corrosionproofing agent for use in the antifreeze incorporates therein at least one member selected from the group consisting of borax, sodium nitrite, phosphoric acid, silicates, sodium benzoate, sodium salt of mercaptobenzothiazole, benzotriazole, methyl benzotriazole, triethylamine, diethanolamine, monoethanolamine, triisopropanolamine, diisopropanolamine, monoisopropanolamine, cyclohexyl amine, ethylenediamine, hydrazine, pyridine, and morpholine (U.S. Patent Nos. 3,046,229; 3,362,910; 3,282,846; 3,046,299; 4,149,985; and 4,333,843). As representatives of these compounds, there can be cited borax, phosphate of triethanolamine, sodium benzoate, sodium nitrite, and sodium silicate. Borax has found popular utility as an effective corrosionproofing agent for engines made of cast iron. In recent years, the general trend toward economization of resources and energies has urged the need of producing automobile parts with lighter materials and has consequently encouraged adoption of aluminum parts. In the circumstances, the fact that borax is deficient in a capacity for corrosionproofing aluminum materials has constituted itself a serious problem.

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It has been known that when an aqueous ethylene glycol solution incorporating borax therein is used in the cooling system of the automobile engine, this solution corrodes aluminum alloy as a material for a cylinder head and a cylinder block in the engine and the product of the corrosion clogs the radiator.

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The phosphate of triethanolamine exhibits an outstanding capacity for corrosionproofing iron and aluminum materials and has found utility as a corrosionproofing agent to take the place of borax. A report has been published which purports that triethanolamine in the presence of a nitrite produces nitrosoamine. It is, therefore, desirable to avoid the occurrence of a nitrite where an amine is being used. The sodium benzoate, when used alone, cannot be expected to manifest a corrosionproofing effect favorably comparable with that of the agents mentioned above.

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The sodium silicate is also effective in curbing the corrosion. It has a disadvantage, however, that it is liable

to undergo separation by gelation during a protracted storage.

An object of this invention, therefore, is to provide a novel antifreeze composition.

Another object of this invention is to provide an antifreeze composition which manifests an outstanding effect in corrosionproofing metal materials.

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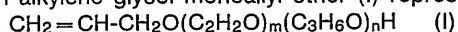
## SUMMARY OF THE INVENTION

The objects described above are accomplished by an antifreeze composition which comprises a glycol and water, contains (A) a phosphoric acid compound, (B) a compound of at least one metal selected from the group consisting of manganese and magnesium, and (C) a corrosionproofing agent, and has a pH value in the range of 6.5 to 9.

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The objects mentioned above are also accomplished by an antifreeze composition which comprises a glycol and water, containing (A) a phosphoric acid compound, (B) a compound of at least one metal selected from the group consisting of manganese and magnesium, (C) a corrosionproofing agent, and (D) at least one compound selected from the group consisting of (a) an aromatic polybasic acid or a salt thereof, (b) an aliphatic dicarboxylic acid or a salt thereof, (c) a molybdate, (d) a tungstate, (e) a homopolymer derived from an alkylene glycol monoallyl ether (I) represented by the formula I:

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wherein m and n are independently 0 or a positive integer, providing that the sum of m and n falls in the range of 1 to 100 and the  $(\text{C}_2\text{H}_4\text{O})$  units and the  $(\text{C}_3\text{H}_6\text{O})$  units may be bound in any order, or a maleic acid type monomer (II) represented by the formula II:

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wherein  $\text{R}^1$  and  $\text{R}^2$  are independently hydrogen atom or methyl group, and X and Y are independently  $(\text{C}_2\text{H}_4\text{O})_p(\text{C}_3\text{H}_6\text{O})_q\text{R}^3$ , wherein  $\text{R}^3$  is alkyl group of 1 to 20 carbon atoms and p and q are independently 0 or a positive integer, providing that the sum of p and q falls in the range of 0 to 100 and the  $(\text{C}_2\text{H}_4\text{O})$  units and the  $(\text{C}_3\text{H}_6\text{O})$  units may be bound in any order, monovalent metal, divalent metal, ammonium group, or organic amine group, and (f) a copolymer derived from the alkylene glycol monoallyl ether (I) or the maleic acid type monomer (II) with a monomer (III) copolymerizable therewith, and has a pH value in the range of 6.5 to 9.

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## DESCRIPTION OF PREFERRED EMBODIMENTS

The glycols which are available for the present invention include ethylene glycol, propylene glycol, butylene glycol, hexylene glycol, and glycerol. They may be used either singly or in the form of a combination of two or more members. The antifreeze which has a main ingredient thereof monoethylene glycol among other compounds cited above is used as added to the cooling liquid in the cooling system for the automobile engine. The weight ratio of the glycol to water is in the range of 99 : 1 to 5 : 95, preferably 98 : 2 to 10 : 90. However, the antifreeze composition is commercially a concentrated state, e.g., the weight ratio of the glycol to water is 99 : 1 to 80 : 20, preferably 98 : 2 to 85 : 5 and is added into the cooling liquid (water) in a certain concentration depending on a freezing temperature.

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The phosphoric acid compounds for use in the present invention are phosphoric acid and salts thereof. Primary through tertiary phosphorus salts of such alkali metals as lithium, sodium, and potassium or ammonia are available for this invention besides orthophosphoric acid. The amount of this compound to be added is in the range of 0.1 to 5% by weight, preferably 0.5 to 3% by weight, based on the amount of the antifreeze as produced.

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Examples of the manganese compound used effectively in this invention include manganese oxide, manganese hydroxide, manganic acid, potassium permanganate, sodium permanganate, lithium permanganate, magnesium permanganate, manganese chromate, manganese borate, manganese fluoride, manganese chloride, manganese bromide, manganese iodide, manganese carbonate, manganese nitrate, manganese sulfite, manganese sulfate, manganese ammonium sulfate, manganese phosphate, manganese hydrogen phosphate, manganese dihydrogen phosphate, manganese ammonium phosphate, manganese formate, manganese acetate, manganese oxalate, manganese butyrate, manganese lactate, manganese malonate, manganese tartrate, manganese succinate, manganese glycerate, manganese valerate, manganese maleate, manganese oleate, manganese citrate, iron manganese citrate, manganese benzoate, manganese salicylate,

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and manganese glutamate. The amount of the manganese compound to be added in this invention is in the range of 0.0001 to 0.05% by weight, preferably 0.005 to 0.02% by weight, based on the amount of the antifreeze as produced.

Examples of the magnesium compound used effectively in this invention include magnesium oxide, magnesium hydroxide, magnesium permanganate, magnesium chromate, magnesium fluoride, magnesium iodide, magnesium carbonate, magnesium borate, magnesium sulfate, magnesium titanate, magnesium tungstate, magnesium borate, magnesium phosphate, magnesium dihydrogen phosphate, magnesium ammonium phosphate, magnesium formate, magnesium acetate, magnesium propionate, magnesium butyrate, magnesium valerate, magnesium laurate, magnesium stearate, magnesium oleate, magnesium glutamate, magnesium lactate, magnesium succinate, magnesium malate, magnesium tartrate, magnesium hydrogen tartrate, magnesium maleate, magnesium citrate, magnesium oxalate, magnesium malonate, magnesium sebacate, magnesium benzoate, magnesium phthalate, magnesium salicylate, and magnesium mandelate. The amount of the magnesium compound to be added in the present invention is in the range of 0.001 to 0.08% by weight, preferably 0.005 to 0.05% by weight, based on the amount of the glycol.

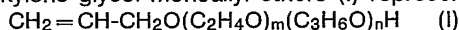
The aromatic polybasic acids available for this invention are benzene-polycarboxyl group substitution products such as phthalic acid, isophthalic acid, terephthalic acid, hemimellitic acid, trimellitic acid, pyromellitic acid. Monovalent metal salts, divalent metal salts, ammonium salts, organic amine salts, and other similar salts of such aromatic polybasic acids are also usable for this invention. The amount of the aromatic polybasic acid to be added in this invention is in the range of 0.05 to 1% by weight, preferably 0.1 to 0.5% by weight, based on the amount of the glycol.

The aliphatic dicarboxylic acids available for the present invention are oxalic acid, malonic acid, succinic acid, glutaric acid, adipic acid, piperic acid, suberic acid, azelaic acid, sebacic acid, brassylic acid, and thapsic acid. Monovalent metal salts, divalent metal salts, ammonium salts, organic amine salts, and other similar salts of these dicarboxylic acid are also usable in this invention. The amount of the dicarboxylic acid to be added herein is in the range of 0.05 to 1% by weight, preferably 0.1 to 0.7% by weight, based on the amount of the glycol.

The molybdates which are used effectively in the present invention include ammonium molybdate, ammonium phosphomolybdate, sodium molybdate, and potassium molybdate, for example. The amount of the molybdate to be added in the present invention is in the range of 0.1 to 1% by weight, preferably 0.3 to 0.7% by weight, based on the amount of the glycol.

The tungstates which are used effectively in the present invention include ammonium tungstate, sodium tungstate, and potassium tungstate, for example. The amount of the tungstate to be added in this invention is in the range of 0.1 to 1% by weight, preferably 0.3 to 0.7% by weight, based on the amount of the glycol.

The polymers which are used effectively in the present invention include monopolymers (e) of polyalkylene glycol monoallyl ethers (I) and/or maleic acid type monomers (II) and copolymers (f) derived from the homopolymers (e) with monomers (III) copolymerizable therewith. Preferably, they are homopolymers of alkylene glycol monoallyl ethers (I) represented by the formula I:



wherein m and n are independently 0 or a positive integer, providing that the sum of m and n falls in the range of 1 to 100, preferably 2 to 50, and the  $(\text{C}_2\text{H}_4\text{O})$  units and the  $(\text{C}_3\text{H}_6\text{O})$  units may be bound in any order, and/or a maleic acid type monomer (II) represented by the formula II:



wherein  $\text{R}^1$  and  $\text{R}^2$  are independently hydrogen atom or methyl group, and X and Y are independently  $(\text{C}_2\text{H}_4\text{O})_p(\text{C}_3\text{H}_6\text{O})_q\text{R}^3$ , wherein  $\text{R}^3$  is alkyl group of 1 to 20 carbon atoms, preferably 1 to 6 carbon atoms and p and q are independently 0 or a positive integer, providing that the sum of p and q falls in the range of 0 to 100, preferably 0 to 50, and the  $(\text{C}_2\text{H}_4\text{O})$  units and the  $(\text{C}_3\text{H}_6\text{O})$  units may be bound in any order, monovalent metal, divalent metal, ammonium group, or organic amine group, and copolymers derived from the homopolymers with monomers (III) copolymerizable therewith.

A polyalkylene glycol monoallyl ether (I) can be synthesized by the conventional method which comprises directly adding ethylene oxide and/or propylene oxide to allyl alcohol in the presence of an alkali such as KOH or NaOH as a catalyst. The compounds represented by the general formulas mentioned above can be used singly or in the form of a combination of two or more members.

The maleic acid type monomers (II) are those represented by the formula II mentioned above. Typical examples of the maleic acid type monomer include maleic acid, phthalic acid, citraconic acid, and mesaconic acid, monovalent metal salts, divalent metal salts, ammonium salts, and organic amine salts of the acids mentioned above, and esters of these acids with alcohols represented by the formula  $\text{HO}(\text{C}_2\text{H}_4\text{O})_p(\text{C}_3\text{H}_6\text{O})_q\text{R}^3$ , wherein  $\text{R}^3$  is hydrogen atom or alkyl group of 1 to 20 carbon atoms, preferably 1 to 6 carbon atoms and p and q are independently 0 or a positive integer, providing that the sum of p and q falls in

the range of 0 to 100, preferably 0 to 50, and the (C<sub>3</sub>H<sub>6</sub>O) units and the (C<sub>2</sub>H<sub>4</sub>O) units may be bound in any order. For example, secondary alcohol ethoxylate monomaleate can be used advantageously. The various maleic acid type monomers (II) cited above may be used singly or in the form of a combination of two or more members.

The monomers (III) which are copolymerizable with the polyalkylene glycol monoallyl ethers (I) and/or the maleic acid type monomers (II) mentioned above are acrylic acid, methacrylic acid, itaconic acid, and crotonic acid, monovalent metal salts, divalent metal salts, ammonium salts, and organic amine salts of the acids mentioned above, esters derived from these acids with alcohols, (meth)acrylamides, vinyl acetate, propenyl acetate, styrene p-methylstyrene, and other aromatic vinyl compounds, and vinyl chloride, for example. These monomers may be used singly or in the form of a combination of two or more members.

The copolymer is derived by using an alkylene glycol monoallyl ether (I), a maleic acid type monomer (II), and/or a monomer (III) copolymerizable therewith in respective proportions of 24 to 75 mol%, 24 to 75 mol%, and 0 to 50 mol%, providing that the total of the components (I), (II), and (III) is 100 mol%.

The polymer is produced by simply polymerizing the component monomers mentioned above with said aid of a polymerization initiator. The polymerization can be carried out in a solvent or by the technique of bulk polymerization.

The polymerization in a solvent can be performed either batchwise or continuously. Examples of the solvent which can be used in the polymerization include water, lower alcohols such as methyl alcohol, ethyl alcohol, and isopropyl alcohol, aromatic or aliphatic hydrocarbons such as benzene, toluene, xylene, cyclohexane, and n-hexane; ethyl acetate; and ketones such as acetone and methylethyl ketone. From the standpoint of solubility of the monomer as raw material and of the polymer as product and conveniences of use of the copolymer, it is desirable to use at least one member selected from the group consisting of lower alcohols having one to four carbon atoms. In the lower alcohols of one to four carbon atoms, methyl alcohol, ethyl alcohol, and isopropyl alcohol are used particularly effectively.

Where the polymerization is carried out in water as a medium, a water-soluble polymerization initiator such as ammonium or alkali metal persulfate or hydrogen peroxide is used. In this case, a promoter such as sodium hydrogen sulfate may be additionally used in the polymerization. In the polymerization using a lower alcohol, an aromatic hydrocarbon, an aliphatic hydrocarbon, ethyl acetate, or a ketone compound as a solvent, a peroxide such as benzoyl peroxide or lauroyl peroxide, a hydroperoxide such as cumene hydroperoxide, or an aliphatic azo compound such as azobisisobutyronitrile is used as a polymerization initiator. Amount of polymerization initiator to be used is in the range of 0.1 to 10 % by weight, based on the monomers. In this case, a promoter such as an amine compound may be additionally used in the polymerization. Where a mixed solvent of water and a lower alcohol is used, a polymerization initiator or a combination of polymerization and promoter suitably selected from among the various compounds cited above may be used. Though the polymerization temperature is decided suitably depending on the kind of the solvent or that of the polymerization initiator to be used, it is generally in the range of 0° to 120°C, preferably 50 to 120 °C.

The bulk polymerization is carried out at a temperature in the range of 50° to 150°C, preferably 80 to 120 °C, in the presence of a polymerization initiator to be selected from among peroxides such as benzoyl peroxide and lauroyl peroxide, hydroperoxides such as cumene hydroperoxide, and aliphatic azo compounds such as azobisisobutyronitrile.

Optionally, the copolymer to be obtained as described above may be neutralized with an alkaline substance before it is put to use. Examples of the alkaline substance usable advantageously for this purpose are hydroxides, chlorides, and carbonates of monovalent metals and divalent metals; ammonia; and organic amines.

Though the molecular weight which the copolymer is required to possess to be effectively used herein falls in a very wide range, it is generally in the range of 500 to 50,000, preferably 1,000 to 30,000.

The copolymer which is derived from a polyalkylene glycol monoallyl ether (I), a maleic acid type monomer (II), and a monomer (III) copolymerizable therewith and is contained in the antifreeze of the present invention manifests an outstanding effect in preventing deposition of scale in addition to an effect in curbing corrosion of iron materials. Thus, the antifreeze of the present invention solves the problem which is suffered by the automobile antifreeze containing a phosphate.

The amount of addition of the copolymer derived from the polyalkylene glycol monoallyl ether (I), the maleic acid type monomer (II), and the monomer (III) copolymerizable therewith is not less than 0.01% by weight, preferably in the range of 0.01 to 0.1 % by weight, based on the amount of the glycol.

The corrosionproofing agent to be added to the antifreeze of the present invention is at least one member selected from the group consisting of sulfites, nitrates, silicates, vanadic acid, vanadates, sodium benzoate, sodium p-tertiary butylbenzoate, sodium mercaptobenzothiazole, methylbenzotriazole, and benzotriazole. As representatives of these corrosionproofing agents, nitrates, sodium mercaptobenzotriazole, methyl benzotriazole, and sodium benzoate can be cited. The amount of the corrosionproofing agent to be added is in the range of 0.05 to 5% by weight, preferably 0.1 to 3% by weight, based on the amount of the glycol.

More specifically, the amount of sodium mercaptobenzothiazole to be added is in the range of 0.05 to 0.7% by weight, preferably 0.1 to 0.5% by weight, based on the amount of the glycol. The nitrate is used in the form of an alkali metal salt such as sodium salt or potassium salt. The amount of the nitrate to be added is in the range of 0.05 to 0.7% by weight, preferably 0.1 to 0.5% by weight, based on the amount of the glycol. The benzoate is used in the form of an alkali metal salt such as sodium salt or potassium salt. The amount of the

benzoate to be added is in the range of 1.0 to 7.0% by weight, preferably 2.0 to 7.0% by weight, based on the amount of the antifreeze as produced. Further, methyl benzotriazole and benzotriazole are also available for this invention.

For the pH adjustment of the antifreeze, any of the basic substances heretofore accepted for the purpose can be used. Preferably, the hydroxide of an alkali metal such as lithium, sodium, or potassium can be used. The pH value of the antifreeze is adjusted in the range of 6.5 to 9, preferably 7 to 8. If the pH value is higher than this range, the antifreeze is deficient in the corrosion proofing to be manifested on aluminum. If the pH value is lower than this range, the antifreeze is capable of corroding iron materials. Thus, the pH value must fall in the range mentioned above to fulfil the effect thereof safely.

The antifreeze of the present invention permits incorporation therein of a defoaming agent such as silicone oil, mineral oil, alcohol, or higher fatty acid ester.

Now, the antifreeze of the present invention will be described more specifically below with reference to working examples and controls. It should be noted, however, that the present invention is not limited to the examples.

A: Method of Test for Metal Corrosion [Japanese Industrial Standard (JIS) K-2234 (Antifreeze) antifreeze concentration 15% by volume]

A test piece of varying metal such as cast aluminum, cast iron, steel, brass, solder, or copper was immersed in a sample antifreeze diluted to 15% by volume with prepared water (obtained by dissolving 148 mg of sodium sulfate, 165 mg of sodium chloride, and 138 mg of sodium hydrogen carbonate in 1 liter of distilled water) and left standing therein at a temperature of  $88 \pm 2^\circ\text{C}$  for 336 hours, with dry air continuously supplied therein at a flow volume of  $100 \pm 10$  ml/min. The mass of the test piece was determined before and after the test to find a change in the mass.

B: Method of Test for corrosion of Heat Transferring Surface

Apparatus -

An apparatus capable of keeping the upper surface of a circular test piece in contact with a sample antifreeze, heating the lower surface of the test piece with a heater, and transferring heat through the test piece to the sample antifreeze was used.

Procedure -

(1) The test piece was polished with a water-resisting abrasive paper, #320, until it acquired flat smooth surface.

(2) A test solution was prepared by diluting a given antifreeze with distilled water or purified water to 20% by volume. To this solution, sodium chloride was added in such an amount that it acquired a chloride ion concentration of 100 ppm.

(3) The sample was placed in the apparatus and then compressed with air to  $0.5 \text{ Kg/cm}^2 \text{ G}$ .

(4) After the test, the test piece was cleaned of adhering precipitate, dried, and weighed accurately.

Test Conditions -

Test piece: Cast aluminum (AC2A)

Temperature of test piece:  $135 \pm 1^\circ\text{C}$

Concentration of antifreeze: Aqueous solution of 20% by volume

Amount of antifreeze: 500 ml

Testing time: 168 hours (continuous)

Test items -

(1) Appearance of test piece

(2) Change in mass of test piece

The change of mass was calculated by the following formula:

$$C = (m_2 - m_1)/X$$

wherein C stands for change in mass ( $\text{mg/cm}^2$ ),  $M_1$  for mass (mg) of the test piece before the test,  $m_2$  for mass (mg) of the test piece after the test, and S for the total surface area ( $\text{cm}^2$ ) of the test piece before the test.

(3) Liquid phase after the test

(4) pH

## C: Method of Determination of Amount of precipitate

In a glass beaker, 400 ml of a sample of varying composition indicated in Table 1 was placed and 400 ml of tap water containing 9 ppm of calcium and 5 ppm of magnesium was added thereto. The beaker was kept at 80°C in an oil bath and left standing therein at rest for 24 hours. The scale formed in the beaker was separated by centrifugation (1500 r.p.m.) to find the amount of precipitate (ml/800 ml). The results were as shown in Table 1.

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## Examples 1 to 6

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Varying components indicated in Table 1 were dissolved in proportions indicated in Table 1 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed. Each of the samples thus prepared was subjected to the following tests and relevant measurements.

A : Test for metal corrosion

B : Test for corrosion of transferring surface

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The results were as shown in Table 1.

## Controls 1 to 5

Samples were prepared by following the procedure of Examples 1 to 6, except that varying components indicated in Table 1 were used instead. They were subjected to the test similarly. The results were as shown in Table 1.

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Table 1

Item	Example					
	1	2	3	4	5	6
Composition of antifreeze (1 by weight)	95	95	95	95	95	95
Monocethylene glycol	5	5	5	5	5	5
Water	1.5		1.5	1.5	1.5	1.5
Phosphoric acid (85% by weight)		2.0				
Potassium Phosphate II						
Phthalic acid	0.3	0.3		0.3	0.3	0.3
Isophthalic acid			0.3			
Magnesium nitrate	0.02	0.02	0.02	0.005	0.05	0.02
Mercaptobenzothiazole	0.3	0.3	0.3	0.3	0.3	0.3
Methyl benzotriazole	0.2	0.2	0.2	0.2	0.2	0.2
Benzotriazole	0.1	0.1	0.1	0.1	0.1	0.1
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	3.0	3.0	3.0	3.0	3.0	3.0
Sodium molybdate	0.5	0.5	0.5	0.5	0.5	0.5
Potassium hydroxide	1.8	0.8	1.8	1.8	1.8	1.0
Sodium hydroxide						
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.02	-0.07	-0.01	-0.13	-0.02	-0.03
Cast iron	-0.03	-0.03	-0.01	-0.12	-0.02	-0.01
Steel	-0.02	-0.01	-0.04	-0.10	-0.03	-0.02
Brass	-0.06	-0.04	-0.05	-0.08	-0.04	-0.04
Solder	-0.07	-0.05	-0.06	-0.10	-0.03	-0.03
Copper	-0.04	-0.03	-0.06	-0.09	-0.05	-0.02
Appearance	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion
Test for corrosion of transferring surface						
change in mass of test piece (mg/cm <sup>2</sup> )	-0.03	-0.05	-0.20	-0.47	-0.00	-0.05
pH before test	7.6	7.6	7.6	7.6	7.6	7.6
pH after test	7.5	7.5	7.7	7.7	7.6	7.6
Appearance of solution after test	not changed	not changed	not changed	not changed	not changed	not changed
Appearance of test piece after test	not changed	not changed	not changed	not changed	not changed	not changed



Table 1 (continued)

Item	Control				
	1	2	3	4	5
Composition of antifreeze (A by weight)	95	95	95	95	95
Monoethylene glycol	5	5	5	5	5
Water					
Phosphoric acid (85% by weight)		1.5	1.5	1.5	1.5
Potassium Phosphate II					
Phthalic acid	0.3		0.3	0.3	0.3
Isophthalic acid					
Magnesium nitrate	0.02	0.02		0.02	0.02
Mercaptobenzothiazole	0.3	0.3	0.3		0.3
Methyl benzotriazole	0.2	0.2	0.2		0.2
Benzotriazole	0.1	0.1	0.1		0.1
Sodium nitrate	0.4	0.4	0.4		0.4
Sodium benzoate	3.0	3.0	3.0		3.0
Sodium molybdate	0.5	0.5	0.5		0.5
Potassium hydroxide	0.8	1.0	1.8	1.8	
Sodium hydroxide					
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-2.23	-0.23	-1.02	-2.81	-0.20
Cast iron	-0.35	-0.08	-0.64	-1.55	-0.58
Steel	-0.25	-0.05	-0.13	-0.42	-0.26
Brass	-0.09	-0.06	-0.11	-0.47	-0.12
Solder	-0.21	-0.08	-0.15	-0.86	-0.10
Copper	-0.14	-0.05	-0.06	-0.56	-0.14
Appearance	Note 1	no corrosion	Note 1	Note 2	Note 3
Test for corrosion of transferring surface					
change in mass of test piece (mg/cm <sup>2</sup> )	-3.15	-2.36	-3.77	-2.20	-1.13
pH before test	7.6	8.0	7.6	7.6	6.2
pH after test	9.5	9.0	8.2	9.3	7.5
Appearance of solution after test	cl	cl	cl	cl	not changed
Appearance of test piece after test	bk	bk	bk	bk	bk

Note 1: cast aluminum and cast iron corrosion

Note 2: cast aluminum, cast iron and solder corrosion

Note 3: cast iron corrosion

cl : cloudy

bk : black

## Referential Example 1 (Preparation of copolymer (1))

In a glass reaction vessel provided with a thermometer, a stirrer, a dropping funnel, a gas inlet tube, and a reflux condenser, 334 parts of polyethylene glycol monoallyl ether (containing an average of five ethylene oxide units per molecule) and 100 parts of water were placed and kept stirred and, after displacement of the inner gas of the reaction vessel with nitrogen, heated to 95°C in an atmosphere of nitrogen. Then, an aqueous solution obtained by dissolving 139.3 parts of maleic acid and 14.2 parts of ammonium persulfate in 225 parts of water was added to the solution in the reaction vessel over a period of 120 minutes. After completion of this addition, 14.2 parts of an aqueous 20% ammonium persulfate solution was added thereto over a period of 20 minutes. After completion of the addition, the inner temperature of the reaction vessel was maintained at 95°C for 100 minutes to complete the polymerization reaction and obtain an aqueous solution of copolymer. This aqueous copolymer solution was neutralized by addition of an aqueous 40% caustic soda solution. Thus, an aqueous solution of a sodium salt of copolymer (1) (hereinafter referred to as "copolymer (1) sodium salt" for short).

This aqueous solution of sodium salt of copolymer (1) had a pH value of 9.5 and a viscosity of 203 cp.

## Referential Example 2 (Preparation of copolymer (2))

In a glass reaction vessel provided with a thermometer, a stirrer, a dropped funnel, a gas inlet tube, and a reflux condenser, 317.3 parts of polyethylene glycol monoallyl ether (containing an average of five ethylene oxide units per molecule) and 88.5 parts of water were placed, kept stirred and, after displacement of the inner gas of the reaction vessel with nitrogen gas, heated to 95°C in an atmosphere of nitrogen. Then, an aqueous solution of 139.3 parts of maleic acid and 11.1 parts of ammonium persulfate in 209 parts of water was added thereto in combination with 2 parts of styrene over a period of 120 minutes. After completion of this addition, 27.8 parts of an aqueous 20% ammonium persulfate solution was added thereto over a period of 60 minutes. After completion of the addition, the inner temperature of the reaction vessel was kept at 95°C for 90 minutes to complete the polymerization and obtained a copolymer (2).

The copolymer (2) was neutralized by addition of an aqueous 40% caustic soda solution, to obtain an aqueous solution of copolymer (2). This aqueous solution had a pH value of 9.5 and a viscosity of 198 cp.

## Referential Example 3 (Preparation of copolymer (3))

In a glass reaction vessel provided with a thermometer, a stirrer, a dropping funnel, a gas inlet tube, and a reflux condenser, 399.6 parts of polyalkylene glycol monoallyl ether (containing 10 ethylene oxide units and two proylene oxide units on the average per molecule) and 203.5 parts of water were placed and kept stirred and, after displacement of the inner gas of the reaction vessel with nitrogen, heated to 95°C in an atmosphere of nitrogen gas. Then, an aqueous solution of 75.4 parts of maleic acid and 17.1 parts of ammonium persulfate in 113.2 parts of water was added thereto over a period of 120 minutes. After completion of this addition, 17.1 parts of an aqueous 20% ammonium persulfate solution was added thereto over a period of 20 minutes. After completion of the addition, the inner temperature of the reaction vessel was maintained at 95°C for 100 minutes to complete the polymerization and produce an aqueous solution of copolymer. Then, this aqueous solution of copolymer was neutralized by addition of an aqueous 40% caustic soda to obtain an aqueous solution of copolymer (3). This solution had a pH value of 9.5 and a viscosity of 135 pc.

## Examples 7 to 14

Varying components indicated in Table 2 were dissolved in varying proportions indicated in Table 2 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

The varying samples were subjected to the following test and relevant measurements.

- A : Test for metal corrosion
- B : Test for corrosion of transferring surface
- C : Determination of amount of precipitate

## Controls 6 to 12

Samples were prepared by following the procedure of Examples 7 to 14, except that varying components indicated in Table 2 were used in varying proportions correspondingly shown in the same table instead. The samples thus obtained were similarly tested. The results were as shown in Table 2.

Table 2

Item	Example													
	7	8	9	10	11	12	13	14						
Composition of antifreeze (1 by weight)	95	95	95	95	95	95	95	95						
Monoethylene glycol	5	5	5	5	5	5	5	5						
Water	1.5	2.0	1.5	1.5	1.5	1.5	1.5	1.5						
Phosphoric acid (85% by weight)	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3						
Potassium Phosphate II	0.007	0.007	0.007	0.007	0.007	0.0005	0.030	0.007						
Phthalic acid	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5						
Isophthalic acid	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3						
Magnesium nitrate	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2						
Sodium molybdate	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1						
Sodium mercaptobenzothiazole	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4						
Methyl benzotriazole	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0						
Benzotriazole	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05						
Sodium nitrate	1.8	0.8	1.8	1.8	1.8	1.8	1.8	1.8						
Sodium benzoate	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00						
Copolymer (1)	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01						
Copolymer (2)	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00						
Copolymer (3)	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01						
Potassium hydroxide	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00						
Sodium hydroxide	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01						
Test for metal corrosion (mg/cm <sup>2</sup> )	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01						
Cast aluminum	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01						
Cast iron	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01						
Steel	-0.02	-0.02	-0.02	-0.02	-0.02	-0.02	-0.02	-0.02						
Brass	-0.02	-0.02	-0.02	-0.02	-0.02	-0.02	-0.02	-0.02						
Solder	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01						
Copper	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01	-0.01						
Appearance	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion						
Amount of precipitate (ml/800ml)	trace	trace	trace	trace	trace	trace	trace	trace						
Test for corrosion of transferring surface	-0.02	-0.04	-0.10	-0.12	-0.12	-0.20	-0.18	-0.07						
change in mass of test piece (mg/cm <sup>2</sup> )	7.7	7.7	7.7	7.6	7.7	7.7	7.7	7.7						
pH before test	7.7	7.7	7.7	7.5	7.7	7.7	7.7	7.7						
pH after test	not	not	not	not	not	not	not	not						
Appearance of solution after test	changed	changed	changed	changed	changed	changed	changed	changed						
Appearance of test piece after test	not	not	not	not	not	not	not	not						
	changed	changed	changed	changed	changed	changed	changed	changed						

Table 2 (continued)

Item	Control										
	6	7	8	9	10	11	12				
Composition of antifreeze (% by weight)	95	95	95	95	95	95	95				
Monoethylene glycol	5	5	5	5	5	5	5				
Water		1.5	1.5	1.5	1.5	1.5	1.5				
Phosphoric acid (85% by weight)											
Potassium Phosphate II											
Phthalic acid	0.3		0.3	0.3	0.3	0.3	0.3				
Isophthalic acid											
Magnesium nitrate	0.007	0.007		0.007	0.007	0.007	0.007				
Sodium molybdate	0.5	0.5	0.5		0.5	0.5	0.5				
Sodium mercaptobenzothiazole	0.3	0.3	0.3	0.3		0.3	0.3				
Methyl benzotriazole	0.2	0.2	0.2	0.2		0.2	0.2				
Benzotriazole	0.1	0.1	0.1	0.1		0.1	0.1				
Sodium nitrate	0.4	0.4	0.4	0.4		0.4	0.4				
Sodium benzoate	3.0	3.0	3.0	3.0		3.0	3.0				
Copolymer (1)	0.05	0.05	0.05	0.05	0.05		0.05				
Copolymer (2)											
Copolymer (3)											
Potassium hydroxide	0.8	1.0	1.8	1.8	1.8	1.8	1.8				
Sodium hydroxide											
Test for metal corrosion (mg/cm <sup>2</sup> )											
Cast aluminum	-1.86	-0.32	-1.20	-2.56	-3.20	-0.03	-0.22				
Steel	-0.34	-0.10	-0.73	-0.65	-1.40	-0.26	-0.36				
Brass	-0.06	-0.03	-0.14	-0.22	-0.18	-0.14	-0.20				
Solder	-0.10	-0.02	-0.10	-0.16	-0.22	-0.06	-0.08				
Copper	-0.64	-0.08	-0.18	-0.22	-1.28	-0.02	-0.01				
Appearance	Note 4	Note 5	Note 1	Note 1	Note 4	Note 6	Note 1				
Amount of precipitate (ml/800ml)	0.08	trace	0.05	trace	0.05	0.50	trace				
Test for corrosion of transferring surface											
change in mass of test piece (mg/cm <sup>2</sup> )	-2.45	-1.40	-4.06	-2.66	-1.45	-0.14	-0.58				
pH before test	7.7	8.2	7.8	7.6	7.7	7.7	6.2				
pH after test	9.6	9.0	8.6	9.2	9.4	7.8	7.3				
Appearance of solution after test	cl	cl	cl	cl	cl	not changed	not changed				
Appearance of test piece after test	bk	bw	bk	bk	bk	not changed	bw				

Note 1 : cast aluminum and cast iron corrosion

Note 4 : squeeze corrosion of cast aluminum and cast iron

Note 5 : squeeze corrosion of cast aluminum

Note 6 : squeeze corrosion of cast iron

bw : blown

cl : cloudy

bk : black

## Examples 15 to 19

Varying components indicated in Table 3 were dissolved in varying proportions indicated in Table 3 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

5

The varying samples thus obtained were subjected to the following test and relevant measurement.  
Test for metal corrosion

The results were as shown in Table 3.

## Controls 13 to 16

10

Varying components indicated in Table 3 were dissolved in varying proportions indicated in Table 3 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

The varying samples thus obtained were subjected to the following test and relevant measurement.

Test for metal corrosion

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The results were as shown in Table 3.

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Table 3

	Example					Control				
	15	16	17	18	19	13	14	15	16	
Composition of antifreeze (% by weight)										
Monethylene glycol	95	95	95	95	95	95	95	95	95	
Water	5	5	5	5	5	5	5	5	5	
Phosphoric acid (8% by weight)	1.5		1.5	1.5	1.5	1.5	1.5	1.5	1.5	
Potassium phosphate II		2.0								
Manganese nitrate	0.007	0.007	0.0005	0.030	0.007		0.007	0.007	0.007	
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	
Sodium benzoate	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	
Potassium hydroxide	1.0	0.4	1.0	1.0		1.0	1.0	1.0	1.0	
Sodium hydroxide					0.5					
Test for metal corrosion (mg/cm <sup>2</sup> )										
Cast aluminum	-0.00	0	-0.01	-0.01	0	-0.68	-0.16	-0.19	-0.18	
Cast iron	-0.01	-0.02	-0.01	-0.03	-0.02	-0.45	-0.18	-0.29	-0.38	
Steel	-0.02	-0.04	-0.01	0	-0.03	-0.33	-0.26	-0.18	-0.18	
Brass	-0.03	-0.01	-0.03	-0.02	0	-0.04	-0.31	-0.06	-0.07	
Solder	-0.04	-0.03	-0.01	-0.02	-0.03	-0.16	-0.26	-0.10	-0.24	
Copper	-0.02	0	-0.01	-0.03	-0.01	-0.02	-0.30	-0.08	0.08	
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	corrosion	corrosion	corrosion	corrosion	

Examples 20 to 25

Varying components indicated in Table 4 were dissolved in varying proportions indicated in Table 4 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

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The varying samples thus obtained were subjected to the following test and relevant measurement.

Test for metal corrosion

The results were as shown in Table 4.

Controls 17 to 21

10

Varying components indicated in Table 4 were dissolved in varying proportions indicated in Table 4 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

The varying samples thus obtained were subjected to the following test and relevant measurement.

Test for metal corrosion

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The results were as shown in Table 4.

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Table 4

	Example					
	20	21	22	23	24	25
Composition of antifreeze (% by weight)						
Monoethylene glycol	95	95	95	95	95	95
Water	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5		1.5	1.5	1.5	1.5
Potassium phosphate II	0.007	2.0				
Magnesium nitrate	0.4	0.007	0.0005	0.030	0.007	0.007
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	5.0	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	0.3	5.0	5.0	5.0	5.0	5.0
Phthalic acid		0.3	0.3	0.3		0.3
Isophthalic acid					0.3	
Potassium hydroxide	1.8	0.8	1.8	1.8	1.8	0.8
Sodium hydroxide						
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.01	-0.06	-0.04	-0.00	-0.02	-0.06
Cast iron	-0.02	-0.03	-0.01	-0.01	-0.06	-0.05
Steel	-0.03	-0.01	-0.04	-0.04	-0.01	-0.00
Brass	-0.00	-0.03	-0.02	-0.06	-0.01	-0.02
Solder	-0.06	-0.09	-0.07	-0.06	-0.08	-0.04
Copper	-0.04	-0.05	-0.03	-0.04	-0.02	-0.04
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion



Table 4 (continued)

	17	18	19	20	21
Composition of antifreeze (% by weight)					
Monoethylene glycol	95	95	95	95	95
Water	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II					
Magnesium nitrate		0.007	0.007	0.007	0.007
Mercaptobenzothiazole soda	0.4		0.4	0.4	0.4
Sodium nitrate	0.4	0.4		0.4	0.4
Sodium benzoate	5.0	5.0	5.0		5.0
Phthalic acid	0.3	0.3	0.3	0.3	
Isophthalic acid					
Potassium hydroxide	1.8	1.8	1.8	1.8	1.0
Sodium hydroxide					
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-1.32	-0.49	-0.96	-0.55	0.64
Cast iron	-0.59	-0.41	-0.46	-1.59	-0.22
Steel	-0.02	-0.29	-0.23	-0.08	-0.06
Brass	-0.06	-0.19	0.01	-0.05	-0.11
Solder	-0.23	-0.30	-0.18	-0.67	-0.23
Copper	-0.08	-0.08	-0.17	-0.14	-0.09
Appearance of test piece	corrosion	corrosion	corrosion	corrosion	corrosion

Examples 26 to 31

5 Varying components indicated in Table 5 were dissolved in varying proportions indicated in Table 5 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

The varying samples thus obtained were subjected to the following test and relevant measurement.  
Test for metal corrosion

The results were as shown in Table 5.

10 Controls 22 to 26

Varying components indicated in Table 5 were dissolved in varying proportions indicated in Table 5 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

15 The varying samples thus obtained were subjected to following test and relevant measurement.

Test for metal corrosion

The results were as shown in Table 5.

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Table 5

	Example				
	26	27	28	29	30
Composition of antifreeze (% by weight)					
Monothylene glycol	95	95	95	95	95
Water	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5		1.5	1.5	1.5
Potassium phosphate II					
Manganese nitrate	0.007	2.0	0.0005	0.030	0.007
Marcaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	5.0	5.0	5.0	5.0	5.0
Sodium molybdate	0.5	0.5	0.5	0.5	0.5
Potassium hydroxide	1.0	0.5	1.0	1.0	1.0
Sodium hydroxide					0.5
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-0.03	-0.02	-0.02	-0.00	-0.03
Cast aluminum	-0.01	-0.05	-0.04	-0.01	-0.04
Steel	-0.01	-0.00	-0.01	-0.01	-0.00
Brass	-0.01	-0.04	-0.02	-0.05	-0.04
Solder	-0.04	-0.08	-0.08	-0.06	-0.05
Copper	-0.01	-0.00	-0.05	-0.02	-0.05
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion

no corrosion

no corrosion

no corrosion

no corrosion

no corrosion

no corrosion

no corrosion

Table 5 (continued)

	Control			
	22	23	24	26
Composition of antifreeze (% by weight)				
Monooethylene glycol	95	95	95	95
Water	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5
Potassium phosphate II				
Manganese nitrate		0.007	0.007	0.007
Mercaptobenzothiazole soda	0.4			
Sodium nitrate	0.4	0.4	0.4	0.4
Sodium benzoate	5.0	5.0	5.0	5.0
Sodium molybdate	0.5	0.5	0.5	0.5
Potassium hydroxide	1.0	1.0	1.0	1.0
Sodium hydroxide				
Test for metal corrosion (mg/cm <sup>2</sup> )				
Cast aluminum	-1.23	-0.55	-0.69	-1.52
Cast aluminum	-0.43	-0.42	-0.54	-0.33
Steel	-0.11	-0.38	-0.29	-0.15
Brass	-0.44	-0.13	-0.35	-0.17
Solder	-0.24	-0.51	-0.23	-0.65
Copper	-0.08	-0.17	-0.18	-0.09
Appearance of test piece	corrosion	corrosion	corrosion	corrosion

Examples 32 to 37

Varying components indicated in Table 6 were dissolved in varying proportions indicated in Table 6 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed. 5  
 The varying samples thus obtained were subjected to the following test and relevant measurement.  
 Test for metal corrosion  
 The results were as shown in Table 6.

Controls 27 to 31 10

Varying components indicated in Table 6 were dissolved in varying proportions indicated in Table 6 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.  
 The varying samples thus obtained were subjected to the following test and relevant measurement. 15  
 Test for metal corrosion  
 The results were as shown in Table 6.

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Table 6

	Example					
	32	33	34	35	36	37
Composition of antifreeze (% by weight)						
Monoethylene glycol	95	95	95	95	95	95
Water	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5		1.5	1.5	1.5	1.5
Potassium phosphate II		2.0				
Manganese nitrate	0.007	0.007		0.030	0.007	0.007
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	5.0	5.0	5.0	5.0	5.0	5.0
Malonic acid	0.5	0.5	0.5	0.5	0.5	0.5
Potassium hydroxide	1.5	1.0	1.5	1.5	1.5	1.0
Sodium hydroxide						
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.02	-0.07	-0.00	-0.04	-0.05	-0.08
Cast iron	-0.02	-0.08	-0.07	-0.09	-0.01	-0.09
Steel	-0.03	-0.01	-0.07	-0.04	-0.08	-0.06
Brass	-0.05	-0.03	-0.08	-0.02	-0.03	-0.06
Solder	-0.08	-0.09	-0.03	-0.04	-0.06	-0.06
Copper	-0.07	-0.06	-0.04	-0.08	-0.08	-0.01
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion

Table 6 (continued)

	Control				
	27	28	29	30	31
Composition of antifreeze (% by weight)					
Monoethylene glycol	95	95	95	95	95
Water	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II					
Manganese nitrate		0.007	0.007	0.007	0.007
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	0.4		0.4	0.4	0.4
Sodium benzoate	5.0	5.0		5.0	5.0
Malonic acid	0.5	0.5	0.5		
Potassium hydroxide	1.5	1.5	1.5	1.5	1.0
Sodium hydroxide					
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-0.86	-0.46	-0.76	-0.62	-1.48
Cast iron	-0.46	-0.34	-0.34	-1.31	-0.88
Steel	-0.08	-0.29	-0.24	-0.12	-0.28
Brass	-0.04	-0.28	-0.09	-0.05	-0.13
Solder	-0.22	-0.25	-0.18	-0.54	-0.27
Copper	-0.04	-0.32	-0.12	-0.13	-0.10
Appearance of test piece	corrosion	corrosion	corrosion	corrosion	corrosion

Examples 38 to 44

Varying components indicated in Table 7 were dissolved in varying proportions indicated in Table 7 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

The varying samples thus obtained were subjected to the following test and relevant measurement.

A : Test for metal corrosion

B : Test for corrosion of the transferring surface

The results were as shown in Table 7.

Controls 32 to 38

Samples were prepared by following the procedure of Example 38 to 44, except that varying components indicated in Table 7 were used in the proportions correspondingly shown in the same table. The samples were similarly tested. The results were as shown in Table 7.



Table 7

	Example						
	38	39	40	41	42	43	44
Composition of antifreeze (% by weight)	95	95	95	95	95	95	95
Monethylene glycol	5	5	5	5	5	5	5
Water	1.5	2.0	1.5	1.5	1.5	1.5	1.5
Phosphoric acid (85% by weight)	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Potassium phosphate II	0.007	0.007	0.007	0.007	0.0005	0.030	0.007
Malonic acid	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Manganese sulfate	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Mercaptothiazole soda	4.0	4.0	4.0	4.0	4.0	4.0	4.0
Sodium nitrate	0.05	0.05	0.05	0.05	0.05	0.05	0.05
Sodium benzoate	1.8	0.8	1.8	1.8	1.8	1.8	1.0
Copolymer (1)							
Copolymer (2)							
Copolymer (3)							
Potassium hydroxide							
Sodium hydroxide							
Test for metal corrosion (mg/cm <sup>2</sup> )							
Cast aluminum	-0.03	-0.08	-0.04	-0.11	-0.04	-0.02	-0.03
Cast iron	-0.02	-0.05	-0.03	-0.10	-0.03	-0.02	-0.05
Steel	-0.04	-0.02	-0.02	-0.09	-0.04	-0.03	-0.05
Brass	-0.06	-0.05	-0.06	-0.08	-0.02	-0.04	-0.09
Solder	-0.06	-0.04	-0.04	-0.11	-0.03	-0.06	-0.06
Copper	-0.04	-0.03	-0.06	-0.09	-0.04	-0.02	-0.04
Appearance	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion
Test for corrosion of heat transferring surface							
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.13	-0.08	-0.18	-0.39	-0.07	-0.15	-0.62
pH before test	7.7	7.7	7.7	7.7	7.7	7.7	7.7
pH after test	7.6	7.5	7.7	7.9	7.7	7.6	7.8
Appearance of liquid after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed
Appearance of test piece after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed

Table 7 (continued)

Composition of antifreeze (% by weight)	Control						
	32	33	34	35	36	37	38
Monoethylene glycol	95	95	95	95	95	95	95
Water	5	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II							
Malonic acid		0.5	0.5	0.5	0.5	0.5	0.5
Manganese sulfate	0.007		0.007	0.007	0.007	0.007	0.007
Mercaptobenzothiazole soda	0.4	0.4		0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4		0.4		0.4
Sodium benzoate	4.0	4.0	4.0	4.0		4.0	4.0
Copolymer (1)	0.05	0.05	0.05	0.05	0.05		0.05
Copolymer (2)							
Copolymer (3)							
Potassium hydroxide	1.0	1.8	1.8	1.8	1.8	1.8	
Sodium hydroxide							
Test for metal corrosion (mg/cm <sup>2</sup> )							
Cast aluminum	-0.43	-2.81	-1.64	-0.48	-0.78	-2.14	-0.22
Cast iron	-0.12	-1.65	-0.88	-1.32	-1.24	-1.08	-0.96
Steel	-0.09	-0.82	-0.46	-0.86	-0.89	-0.76	-0.53
Brass	-0.11	-0.48	-0.92	-0.08	-0.08	-0.13	-0.06
Solder	-0.15	-0.86	-0.68	-0.22	-0.94	-0.58	-0.16
Copper	-0.08	-0.46	-0.79	-0.06	-0.06	-0.11	-0.07
Appearance	Note 5	Note 1	Note 1*	Note 1	Note 2	Note 2	Note 7
Test for corrosion of heat transferring surface							
Change in mass of test piece (mg/cm <sup>2</sup> )	-1.08	-2.08	-1.20	-0.89	-0.54	-1.03	-0.42
pH before test	7.9	7.7	7.6	7.7	7.7	7.7	6.2
pH after test	9.2	8.9	8.8	8.2	7.9	8.6	7.3
Appearance of liquid after test	cl	cl	cl	cl	not changed	cl	not changed
Appearance of test piece after test	bk	bk	bk	bk	not changed	bk	bw

Note 1 : cast aluminum and cast iron corrosion

Note 2 : cast aluminum, cast iron and solder corrosion

Note 5 : squeezeout corrosion of cast aluminum

Note 7 : cast iron and steel corrosion

\* : copperchanged color

cl : cloudy

bk : black

bw : brown

## Example 45 to 51

Varying components indicated in Table 8 were dissolved in varying proportions indicated in Table 8 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

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The varying samples thus obtained were subjected to the following test and relevant measurement.

A Test for metal corrosion

B Test for corrosion of transferring surface.

The results were as shown in Table 8.

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## Controls 39 to 46

Samples were prepared by following the procedure of Examples 45 to 51, except that varying components indicated in Table 8 were used in the proportions correspondingly shown in the same table. The samples are similarly tested. The results were as shown in Table 8.

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Table 8

	Example					
	45	46	47	48	49	50
Composition of antifreeze (% by weight)						
Monethylene glycol	95	95	95	95	95	95
Water	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II						
Malonic acid	0.5	2.0	0.5	0.5	0.5	0.5
Manganese nitrate	0.007	0.007	0.007	0.007	0.0005	0.007
Sodium molybdate	0.5	0.5	0.5	0.5	0.5	0.5
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	4.0	4.0	4.0	4.0	4.0	4.0
Copolymer (1)	0.05	0.05			0.05	0.05
Copolymer (2)			0.05			
Copolymer (3)				0.05		
Potassium hydroxide	1.8	0.8	1.8	1.8	1.8	1.8
Sodium hydroxide						1.0
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.01	-0.04	-0.03	-0.06	-0.02	-0.02
Cast iron	-0.02	-0.03	-0.04	-0.06	-0.03	-0.03
Steel	-0.03	-0.01	-0.01	-0.05	-0.02	-0.04
Brass	-0.04	-0.03	-0.03	-0.03	-0.03	-0.04
Solder	-0.03	-0.02	-0.02	-0.08	-0.06	-0.06
Copper	-0.02	-0.02	-0.03	-0.05	-0.02	-0.03
Appearance	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion
Test for corrosion of heat						
Transferring surface						
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.03	-0.06	-0.08	-0.13	-0.04	-0.08
pH before test	7.7	7.7	7.7	7.7	7.7	7.7
pH after test	7.6	7.5	7.7	7.9	7.7	7.6
Appearance of liquid after test	not changed	not changed	not changed	not changed	not changed	not changed
Appearance of test piece after test	not changed	not changed	not changed	not changed	not changed	not changed

Table 8 (continued)

	Control									
	39	40	41	42	43	44	45	46		
Composition of antifreeze (% by weight)										
Monethylene glycol	95	95	95	95	95	95	95	95		
Water	5	5	5	5	5	5	5	5		
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5		
Potassium phosphate II										
Malonic acid		0.5	0.5	0.5	0.5	0.5	0.5	0.5		
Manganese nitrate	0.007		0.007	0.007	0.007	0.007	0.007	0.007		
Sodium molybdate	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5		
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4		
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4		
Sodium benzoate	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0		
Copolymer (1)	0.05	0.05	0.05	0.05	0.05	0.05				
Copolymer (2)										
Copolymer (3)										
Potassium hydroxide	1.0	1.8	1.8	1.8	1.8	1.8	1.5			
Sodium hydroxide										
Test for metal corrosion (mg/cm <sup>2</sup> )										
Cast alum	-0.32	-1.41	-0.96	-0.32	-0.48	-1.07	-0.42	-0.36		
Cast iron	-0.18	-0.83	-0.51	-0.86	-0.76	-0.53	-0.83	-0.88		
Steel	-0.06	-0.48	-0.32	-0.42	-0.29	-0.38	-0.38	-0.46		
Brass	-0.08	-0.26	-0.42	-0.08	-0.06	-0.09	-0.06	-0.09		
Solder	-0.13	-0.43	-0.34	-0.18	-0.47	-0.29	-0.16	-0.26		
Copper	-0.06	-0.23	-0.36	-0.04	-0.04	-0.06	-0.11	-0.08		
Appearance	Note 5	Note 1	Note 1*	Note 1	Note 2	Note 2	Note 1	Note 7		
Test for corrosion of heat transferring surface										
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.78	-1.04	-1.02	-0.69	-0.46	-0.98	-0.11	-0.48		
pH before test	7.9	7.7	7.6	7.7	7.7	7.7	7.7	6.4		
pH after test	8.5	8.3	7.9	8.1	8.0	8.7	8.8	7.5		
Appearance of liquid after test	cl	cl	cl	cl	not changed	cl	cl	not changed		
Appearance of test piece after test	bk	bk	bk	bk	not changed	bk	gray	bw		

Note 1 : cast aluminum and cast iron corrosion

Note 2 : cast aluminum, cast iron and solder corrosion

Note 5 : squeezeout corrosion of cast aluminum

Note 7 : cast iron and steel corrosion

\* : copper changed color

cl : cloudy

bk : black

bw : brown

Examples 52 to 58

5 Varying components indicated in Table 9 were dissolved in varying proportions indicated in Table 9 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

The varying samples thus obtained were subjected to the following test and relevant measurement.

A : Test for metal corrosion

B : Test for corrosion of transferring surface The results were as shown in Table 9.

10 Controls 47 to 51

Samples were prepared by following the procedure of Examples 52 to 58, except that varying components indicated in Table 9 were used in the proportions correspondingly shown in the same table. The samples were similarly tested. The results were as shown in Table 9.

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Table 9

	Example						
	52	53	54	55	56	57	58
Composition of antifreeze (% by weight)	95	95	95	95	95	95	95
Monoethylene glycol	5	5	5	5	5	5	5
Water	1.5	2.0	1.5	1.5	1.5	1.5	1.5
Phosphoric acid (85% by weight)	0.3		0.3	0.3	0.3	0.3	0.3
Potassium phosphate II							
Phthalic acid	0.007	0.3	0.007	0.007	0.0005	0.030	0.007
Isophthalic acid	0.3	0.007	0.007	0.007	0.3	0.3	0.3
Manganese nitrate	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Mercapto benzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	4.0	4.0	4.0	4.0	4.0	4.0	4.0
Sodium benzoate	0.05	0.05			0.05	0.05	0.05
Copolymer (1)			0.05	0.05			
Copolymer (2)							
Copolymer (3)							
Potassium hydroxide	1.8	0.8	1.8	1.8	1.8	1.8	1.0
Sodium hydroxide							
Test for metal corrosion (mg/cm <sup>2</sup> )							
Cast aluminum	-0.04	-0.02	-0.01	-0.07	-0.02	-0.01	-0.07
Cast iron	-0.03	-0.03	-0.04	-0.05	-0.02	-0.01	-0.09
Steel	-0.04	-0.02	-0.01	-0.07	-0.02	-0.04	-0.05
Brass	-0.05	-0.00	-0.04	-0.04	-0.03	-0.03	-0.04
Solder	-0.06	-0.02	-0.04	-0.10	-0.09	-0.08	-0.11
Copper	-0.03	-0.02	-0.01	-0.05	-0.04	-0.01	-0.06
Appearance	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion
Test for corrosion of heat transferring surface							
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.14	-0.06	-0.22	-0.32	-0.13	-0.28	-0.42
pH before test	7.7	7.7	7.7	7.7	7.7	7.7	7.7
pH after test	7.7	7.5	7.8	8.0	7.5	7.6	8.0
Appearance of liquid after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed
Appearance of test piece after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed

Table 9 (continued)

	Control				
	47	48	49	50	51
Composition of antifreeze (% by weight)					
Monoethylene glycol	95	95	95	95	95
Water	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II					
Phthalic acid	0.3	0.3	0.3	0.3	0.3
Isophthalic acid					
Manganese nitrate	0.007		0.007	0.007	0.007
Mercaptobenzothiazole soda	0.3	0.3		0.3	0.3
Sodium nitrate	0.4	0.4		0.4	0.4
Sodium benzoate	4.0	4.0		4.0	4.0
Copolymer (1)	0.05	0.05	0.05		0.05
Copolymer (2)					
Copolymer (3)					
Potassium hydroxide	1.0	1.8	1.8	1.8	
Sodium hydroxide					
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-0.52	-2.14	-1.86	-0.82	-0.28
Cast iron	-0.22	-0.88	-1.08	-1.09	-0.68
Steel	-0.11	-0.38	-0.52	-0.30	-0.32
Brass	-0.08	-0.26	-0.38	-0.08	-0.06
Solder	-0.14	-0.33	-0.45	-0.37	-0.18
Copper	-0.07	-0.21	-0.36	-0.04	-0.04
Appearance	Note 5	Note 1	Note 2*	Note 1	Note 7
Test for corrosion of heat transferring surface					
Change in mass of test piece (mg/cm <sup>2</sup> )	-1.35	-3.12	-1.02	-0.69	-0.46
pH before test	7.9	7.7	7.6	7.7	6.3
pH after test	9.0	9.3	7.9	8.1	8.0
Appearance of liquid after test	cl	cl	cl	cl	not changed
Appearance of test piece after test	bk	bk	bk	bk	not changed

Note 1 : cast aluminum and cast iron corrosion

Note 2 : cast aluminum, cast iron and solder corrosion

Note 5 : squeezeout corrosion of cast aluminum

Note 7 : cast iron and steel corrosion

\* : copper changed color

cl : cloudy

bk : black

bw : brown



## Examples 59 to 65

Varying components indicated in Table 10 were dissolved in varying proportions indicated in Table 10 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed. 5

The varying samples thus obtained were subjected to the following test and relevant measurement.  
Test for metal corrosion

The results were as shown in Table 10.

## Controls 52 to 55

Samples were prepared by following the procedure of Examples 59 to 65, except that varying components indicated in Table 10 were used in the proportions correspondingly shown in the same table. The samples were similarly tested. The results were as shown in Table 10. 10

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Table 10

	Example					
	59	60	61	62	63	64
Composition of antifreeze (% by weight)						
Diethylene glycol	95	95	95	95	95	95
Water	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	2.0	1.5	1.5	1.5	1.5
Potassium phosphate II	0.007	0.007	0.007	0.007	0.0005	0.030
Manganese nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Macrocaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	4.0	4.0	4.0	4.0	4.0	4.0
Sodium benzoate	0.05	0.05	0.05	0.05	0.05	0.05
Copolymer (1)						
Copolymer (2)						
Copolymer (3)						
Potassium hydroxide	1.0	0.4	1.0	1.0	1.0	1.0
Sodium hydroxide						
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.02	-0.04	-0.04	-0.08	-0.07	-0.09
Cast aluminum	-0.04	-0.07	-0.09	-0.05	-0.05	-0.09
Steel	-0.08	-0.00	-0.04	-0.06	-0.02	-0.06
Brass	-0.08	-0.09	-0.01	-0.05	-0.02	-0.03
Solder	-0.08	-0.09	-0.01	-0.08	-0.05	-0.07
Copper	-0.07	-0.06	-0.01	-0.06	-0.06	-0.03
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion

Table 10 (continued)

	Control			
	52	53	54	55
Composition of antifreeze (% by weight)				
Moethylene glycol Water	95	95	95	95
Water	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5
Potassium phosphate II				
Manganese nitrate		0.007	0.007	0.007
Macrcaptobenzothiazole soda	0.4		0.4	0.4
Sodium nitrate	0.4		0.4	0.4
Sodium benzoate	4.0		4.0	4.0
Copolymer (1)	0.05	0.05		0.5
Copolymer (2)				
Copolymer (3)				
Potassium hydroxide	1.0	1.0	1.0	
Sodium hydroxide				
Test for metal corrosion (mg/cm <sup>2</sup> )				
Cast aluminum	-2.14	-1.14	-0.88	-0.24
Steel	-0.16	-1.61	-1.57	-0.56
Brass	-0.65	-0.61	-0.29	-0.28
Solder	-0.12	-0.29	-0.10	-0.06
Copper	-0.23	-0.45	-0.28	-0.21
Appearance of test piece	-0.15	-0.48	-0.08	-0.08
	corrosion	corrosion	corrosion	corrosion

Examples 66 to 73

5 Varying components indicated in Table 11 were dissolved in varying proportions indicated in Table 11 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

The varying samples thus obtained were subjected to the following test and relevant measurement.

A : Test for metal corrosion

B : Test for corrosion of transferring surface

The results were as shown in Table 11.

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Controls 56 to 62

15 Samples were prepared by following the procedure of Example 66 to 73, except that varying components indicated in Table 11 were used in the proportions correspondingly shown in the same table. The samples were similarly tested. The results were as shown in Table 11.

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Table 11

Item	Example									
	66	67	68	69	70	71	72	73		
Composition of antifreeze (A by weight)	95	95	95	95	95	95	95	95		
Monoethylene glycol	5	5	5	5	5	5	5	5		95
Water	1.5	2.0	1.5	1.5	1.5	1.5	1.5	1.5		5
Phosphoric acid (85% by weight)										1.5
Potassium Phosphate II	0.3	0.3		0.3	0.3	0.3	0.3	0.3		0.3
Phthalic acid										
Isophthalic acid	0.02	0.02	0.3	0.02	0.02	0.005	0.05	0.02		0.02
Magnesium nitrate	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5		0.5
Sodium molybdate	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3		0.3
Sodium mercaptobenzothiazole	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2		0.2
Methyl benzotriazole	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1		0.1
Benzotriazole	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4		0.4
Sodium nitrate	3.0	3.0	3.0	3.0	3.0	3.0	3.0	3.0		3.0
Sodium benzoate	0.05	0.05	0.05	0.05		0.05	0.05	0.05		0.05
Copolymer (1)										
Copolymer (2)					0.05					
Copolymer (3)										
Potassium hydroxide	1.8	0.8	1.8	1.8	1.8	1.8	1.8	1.8		0.8
Sodium hydroxide										
Test for metal corrosion (mg/cm <sup>2</sup> )										
Cast aluminum	-0.06	-0.04	-0.02	-0.06	-0.07	-0.03	-0.07	-0.04		-0.04
Cast iron	-0.02	-0.06	-0.05	-0.04	-0.04	-0.05	-0.03	-0.05		-0.02
Steel	-0.06	-0.05	-0.07	-0.01	-0.01	-0.03	-0.03	-0.03		-0.06
Brass	-0.06	-0.04	-0.02	-0.04	-0.00	-0.05	-0.04	-0.03		-0.03
Solder	-0.04	-0.05	-0.03	-0.06	-0.06	-0.01	-0.00	-0.00		-0.00
Copper	-0.01	-0.03	-0.00	-0.00	-0.00	-0.00	-0.06	-0.06		-0.06
Appearance	corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion		no corrosion
Test for corrosion of transferring surface										
change in mass of test piece (mg/cm <sup>2</sup> )	-0.04	-0.06	-0.09	-0.13	-0.12	-0.18	-0.20	-0.07		-0.07
pH before test	7.8	7.8	7.8	7.8	7.8	7.8	7.8	7.8		7.8
pH after test	7.8	7.8	7.8	7.7	7.8	7.8	7.8	7.8		7.8
Appearance of solution after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed	not changed		not changed
Appearance of test piece after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed	not changed		not changed

Table 11 (continued)

Item	Control					
	56	57	58	59	60	61
Composition of antifreeze (1 by weight)						
Monothylene glycol	95	95	95	95	95	95
Water	5	5	5	5	5	5
Phosphoric acid (85% by weight)		1.5	1.5	1.5	1.5	1.5
Potassium Phosphate II						
Phthalic acid	0.3		0.3	0.3	0.3	0.3
Isophthalic acid						
Magnesium nitrate	0.02	0.02		0.02	0.02	0.02
Sodium molybdate	0.5	0.5	0.5		0.5	0.5
Sodium mercaptobenzothiazole	0.3	0.3	0.3	0.3	0.3	0.3
Methyl benzotriazole	0.2	0.2	0.2	0.2	0.2	0.2
Benztotriazole	0.1	0.1	0.1	0.1	0.1	0.1
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	3.0	3.0	3.0	3.0	3.0	3.0
Copolymer (1)	0.05	0.05	0.05	0.05		
Copolymer (2)						
Copolymer (3)						
Potassium hydroxide	0.8	1.0	1.8	1.8	1.8	1.8
Sodium hydroxide						
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-1.61	-0.98	-1.38	-2.20	-3.66	-0.08
Cast iron	-0.20	-0.19	-0.50	-0.64	-1.16	-0.25
Steel	-0.07	-0.02	-0.13	-0.54	-0.19	-0.13
Brass	-0.19	-0.02	-0.16	-0.11	-0.21	-0.06
Solder	-0.82	-0.02	-0.19	-0.22	-1.12	-0.09
Copper	-0.04	-0.06	-0.02	-0.16	-0.16	-0.02
Appearance	Note 4	Note 5	Note 1	Note 1	Note 4	Note 6
Test for corrosion of transferring surface						
change in mass of test piece (mg/cm <sup>2</sup> )	-2.67	-1.54	-4.45	-2.26	-1.69	-0.12
pH before test	7.8	8.2	7.9	7.7	7.8	7.8
pH after test	9.8	9.2	8.5	9.4	9.2	7.4
Appearance of solution after test	cl	cl	cl	cl	cl	not changed
Appearance of test piece after test	bk	bw	bk	bk	bk	not changed

Note 1 : cast aluminum and cast iron corrosion

Note 4 : squeeze-out corrosion of cast aluminum and cast iron

Note 5 : squeeze-out corrosion of cast aluminum

Note 6 : squeeze-out corrosion of cast iron

bw : blown

cl : cloudy

bk : black

## Examples 67 to 89

Varying components indicated in Tables 12 through 15 were dissolved in varying proportions indicated in Tables 12 through 15 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed. 5

The varying samples thus obtained were subjected to the following test and relevant measurement.

Test for metal corrosion

The results were as shown in Tables 12 through 15. 10

## Controls 63 to 81

Varying components indicated in Tables 12 through 15 were dissolved in varying proportions indicated in Tables 12 through 15 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed. 15

The varying samples thus obtained were subjected to the following test and relevant measurement.

Test for metal corrosion

The results were as shown in Tables 12 through 15. 20

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Table 12

	Example					
	67	68	69	70	71	72
Composition of antifreeze						
(% by weight)	95	95	95	95	95	95
Monoethylene glycol	5	5	5	5	5	5
Water	1.5		1.5	1.5	1.5	1.5
Phosphoric acid (85% by weight)		2.0				
Potassium phosphate II	0.02	0.02	0.005	0.05	0.02	0.02
Magnesium nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	5.0	5.0	5.0	5.0	5.0	5.0
Sodium benzoate	0.3	0.3	0.3	0.3	0.3	0.3
Phthalic acid						
Isophthalic acid	1.8	1.2	1.8	1.8	1.8	
Potassium hydroxide						0.8
Sodium hydroxide						
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.03	-0.05	-0.01	-0.05	-0.06	-0.01
Cast iron	-0.03	-0.06	-0.04	-0.05	-0.01	-0.02
Steel	-0.05	-0.05	-0.06	-0.06	-0.06	-0.03
Brass	-0.03	-0.01	-0.03	-0.02	-0.06	-0.02
Solder	-0.05	-0.01	-0.01	-0.03	-0.01	-0.06
Copper	-0.04	-0.05	-0.05	-0.05	-0.06	-0.02
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion



Table 12 (continued)

	Control				
	63	64	65	66	67
Composition of antifreeze (% by weight)					
Monocethylene glycol	95	95	95	95	95
Water	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II					
Magnesium nitrate	0.4	0.02	0.02	0.02	0.02
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	5.0	5.0	5.0	5.0	5.0
Sodium benzoate	0.3	0.3	0.3	0.3	0.3
Phthalic acid					
Isophthalic acid	1.8	1.8	1.8	1.8	1.8
Potassium hydroxide					
Sodium hydroxide					
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-1.27	-0.86	-0.51	-0.13	-0.94
Cast iron	-0.13	-0.57	-0.40	-1.26	-0.21
Steel	-0.08	-0.30	-0.27	-0.10	-0.06
Brass	-0.04	-0.13	-0.09	-0.06	-0.13
Solder	-0.23	-0.32	-0.22	-0.55	-0.19
Copper	-0.06	-0.18	-0.04	-0.13	-0.08
Appearance of test piece	corrosion	corrosion	corrosion	corrosion	corrosion

Table 13

	Example					
	73	74	75	76	77	78
Composition of antifreeze (% by weight)						
Monoethylene glycol	95	95	95	95	95	95
Water	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5		1.5	1.5	1.5	1.5
Potassium phosphate II		2.0				
Magnesium nitrate	0.02	0.02	0.005	0.05	0.02	0.02
Marcaptobenzoethiazole soda	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	5.0	5.0	5.0	5.0	5.0	5.0
Sodium molybdate	0.5	0.5	0.5	0.5	0.5	0.5
Potassium hydroxide	1.0	0.5	1.0	1.0	1.0	0.5
Sodium hydroxide						
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.04	-0.04	-0.04	-0.00	-0.04	-0.00
Cast aluminum	-0.01	-0.00	-0.03	-0.02	-0.05	-0.03
Steel	-0.02	-0.05	-0.05	-0.05	-0.04	-0.04
Brass	-0.04	-0.02	-0.04	-0.02	-0.01	-0.00
Solder	-0.04	-0.05	-0.02	-0.04	-0.02	-0.05
Copper	-0.01	-0.04	-0.01	-0.02	-0.00	-0.00
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion

Table 13 (continued)

	Control				
	68	69	70	71	72
Composition of antifreeze (% by weight)					
Monoethylene glycol	95	95	95	95	95
Water	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II					
Magnesium nitrate		0.02	0.02	0.02	0.02
Marcaptobenzothiazole soda	0.4		0.4	0.4	0.4
Sodium nitrate	0.4	0.4		0.4	
Sodium benzoate	5.0	5.0	5.0		5.0
Sodium molybdate	0.5	0.5	0.5	0.5	
Potassium hydroxide	1.0	1.0	1.0	1.0	1.0
Sodium hydroxide					
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-1.22	-0.82	-0.51	-0.48	-1.11
Cast aluminum	-0.24	-0.47	-0.40	-1.30	-0.31
Steel	-0.10	-0.38	-0.31	-0.09	-0.08
Brass	-0.24	-0.13	-0.29	-0.16	-0.18
Solder	-0.33	-0.42	-0.22	-0.45	-0.49
Copper	-0.06	-0.18	-0.24	-0.13	-0.08
Appearance of test piece	corrosion	corrosion	corrosion	corrosion	corrosion

Table 14

	Example					
	79	80	81	82	83	84
Composition of antifreeze (% by weight)						
Monoethylene glycol	95	95	95	95	95	95
Water	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5		1.5	1.5	1.5	1.5
Potassium phosphate II		2.0				
Magnesium nitrate	0.02	0.02	0.005	0.05	0.02	0.02
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	5.0	5.0	5.0	5.0	5.0	5.0
Malonic acid	0.5	0.5	0.5	0.5	0.5	0.5
Potassium hydroxide	1.5	0.7	1.5	1.5	1.5	
Sodium hydroxide						0.7
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.08	-0.02	-0.04	-0.03	-0.02	-0.08
Cast iron	-0.07	-0.04	-0.03	-0.07	-0.07	-0.02
Steel	-0.06	-0.03	-0.00	-0.08	-0.02	-0.08
Brass	-0.04	-0.05	-0.00	-0.06	-0.03	-0.00
Solder	-0.01	-0.03	-0.03	-0.05	-0.00	-0.01
Copper	-0.08	-0.01	-0.07	-0.05	-0.06	-0.02
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion

Table 14 (continued)

	Control				
	73	74	75	76	77
Composition of antifreeze (% by weight)					
Monoethylene glycol	95	95	95	95	95
Water	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II					
Magnesium nitrate		0.02	0.02	0.02	0.02
Mercaptobenzothiazole soda	0.4		0.4	0.4	0.4
Sodium nitrate	0.4	0.4		0.4	0.4
Sodium benzoate	5.0	5.0	5.0		5.0
Malonic acid	0.5	0.5	0.5	0.5	
Potassium hydroxide	1.5	1.5	1.5	1.5	1.0
Sodium hydroxide					
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-0.84	-0.46	-0.56	-0.56	-1.48
Cast iron	-0.44	-0.37	-0.44	-1.30	-0.51
Steel	-0.07	-0.28	-0.21	-0.13	-0.18
Brass	-0.04	-0.23	-0.09	-0.06	-0.12
Solder	-0.23	-0.32	-0.20	-0.43	-0.23
Copper	-0.06	-0.28	-0.14	-0.13	-0.08
Appearance of test piece	corrosion	corrosion	corrosion	corrosion	corrosion

Table 15

	Example					Control				
	85	86	87	88	89	78	79	80	81	
Composition of antifreeze (% by weight)										
Monothylene glycol	95	95	95	95	95	95	95	95	95	
Water	5	5	5	5	5	5	5	5	5	
Phosphoric acid (85% by weight)	1.5		1.5	1.5	1.5	1.5	1.5	1.5	1.5	
Potassium phosphate II		2.0								
Magnesium nitrate	0.02	0.02	0.005	0.05	0.02		0.02	0.02	0.02	
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	
Sodium benzoate	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	5.0	
Potassium hydroxide	1.0	0.5	1.0	1.0		1.0	1.0	1.0	1.0	
Sodium hydroxide					0.5					
Test for metal corrosion (mg/cm <sup>2</sup> )										
Cast aluminum	-0.02	-0.02	-0.01	-0.04	-0.01	-0.79	-0.52	-0.28	-0.32	
Cast iron	-0.02	-0.03	-0.00	-0.05	-0.01	-0.58	-0.13	-0.32	-0.40	
Steel	-0.00	-0.01	-0.00	-0.05	-0.05	-0.33	-0.28	-0.20	-0.17	
Brass	-0.05	-0.03	-0.01	-0.05	-0.00	-0.05	-0.34	-0.03	-0.09	
Solder	-0.04	-0.04	-0.03	-0.03	-0.00	-0.21	-0.30	-0.12	-0.22	
Copper	-0.00	-0.05	-0.01	-0.03	-0.02	-0.02	-0.26	-0.06	-0.04	
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	corrosion	corrosion	corrosion	corrosion	

## Examples 90 to 110

Varying components indicated in Tables 16 through 19 were dissolved in varying proportions indicated in Tables 16 through 19 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed. 5

The varying samples thus obtained were subjected to the following test and relevant measurement.

A : Test for metal corrosion

B : Test for corrosion of transferring surface

The results were as shown in Table 7. 10

## Controls 82 to 101

Samples were prepared by following the procedure of Examples 90 to 110, except that varying components indicated in Tables 16 through 18 were used in the proportions correspondingly shown in the same table. The samples were similarly tested. The results were as shown in Tables 16 through 18. 15

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Table 16

	Example					
	90	91	92	93	94	95
Composition of antifreeze (% by weight)						
Monethylene glycol	95	95	95	95	95	95
Water	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5		1.5	1.5	1.5	1.5
Potassium phosphate II		2.0				
Malonic acid	0.5	0.5	0.5	0.5	0.5	0.5
Magnesium nitrate	0.02	0.02	0.02	0.02	0.005	0.05
Sodium molybdate	0.5	0.5	0.5	0.5	0.5	0.5
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	4.0	4.0	4.0	4.0	4.0	4.0
Copolymer (1)	0.05	0.05			0.05	0.05
Copolymer (2)			0.05			
Copolymer (3)				0.05		
Potassium hydroxide	1.8	0.8	1.8	1.8	1.8	1.8
Sodium hydroxide						
Test for metal corrosion (mg/cm <sup>2</sup> )						
Cast aluminum	-0.02	-0.00	-0.02	-0.01	-0.02	-0.04
Cast iron	-0.03	-0.04	-0.02	-0.01	-0.03	-0.04
Steel	-0.03	-0.02	-0.00	-0.01	-0.00	-0.04
Brass	-0.01	-0.00	-0.03	-0.01	-0.01	-0.01
Solder	-0.04	-0.04	-0.03	-0.04	-0.02	-0.03
Copper	-0.02	-0.00	-0.03	-0.04	-0.03	-0.04
Appearance	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion
Test for corrosion of heat transferring surface						
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.02	-0.08	-0.07	-0.03	-0.07	-0.10
pH before test	7.8	7.8	7.8	7.8	7.8	7.8
pH after test	7.8	7.8	7.8	7.8	7.6	7.8
Appearance of liquid after test	not changed	not changed	not changed	not changed	not changed	not changed
Appearance of test piece after test	not changed	not changed	not changed	not changed	not changed	not changed



Table 16 (continued)

	82	83	84	85	86	87	88	89
Composition of antifreeze (% by weight)	95	95	95	95	95	95	95	95
monoethylene glycol	5	5	5	5	5	5	5	5
Water	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Phosphoric acid (85% by weight)								
Potassium phosphate II								
Malonic acid	0.02	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Magnesium nitrate	0.5	0.5	0.02	0.02	0.02	0.02	0.02	0.2
Sodium molybdate	0.4	0.4	0.4	0.5	0.5	0.5	0.5	0.5
Mercaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	4.0	4.0	4.0	4.0	4.0	4.0	4.0	4.0
Sodium benzoate								
Copolymer (1)	0.05	0.05	0.05	0.05	0.05	0.05		0.05
Copolymer (2)								
Copolymer (3)								
Potassium hydroxide	1.0	1.8	1.8	1.8	1.8	1.8		
Sodium hydroxide								
Test for metal corrosion (mg/cm <sup>2</sup> )								
Cast aluminum	-0.34	-1.46	-0.99	-0.38	-0.52	-1.68	-0.38	-0.20
Cast iron	-0.12	-0.78	-0.49	-0.60	-0.64	-0.66	-0.65	-0.31
Steel	-0.08	-0.47	-0.42	-0.13	-0.24	-0.29	-0.43	-0.24
Brass	-0.11	-0.29	-0.42	-0.06	-0.11	-0.11	-0.06	-0.06
Solder	-0.18	-0.22	-0.22	-0.19	-0.42	-0.60	-0.19	-0.04
Copper	-0.06	-0.24	-0.46	-0.06	-0.09	-0.06	-0.09	-0.12
Appearance	Note 5	Note 1	Note 1*	Note 1	Note 2	Note 2	Note 1	Note 7
Test for corrosion of heat transferring surface								
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.58	-1.67	-2.34	-1.45	-1.26	-1.29	-0.18	-0.79
pH before test	7.8	7.8	8.2	7.9	7.7	7.8	7.8	6.4
pH after test	8.8	9.2	9.2	8.5	9.4	9.0	7.8	7.4
Appearance of liquid after test	not changed	cl	cl	cl	cl	cl	not changed	not changed
Appearance of test piece after test	not changed	bk	bw	bk	bk	bk	not changed	bw

Note 1 : cast aluminum and cast iron corrosion

Note 2 : cast aluminum, cast iron and solder corrosion

Note 5 : squeezeout corrosion of cast aluminum

Note 7 : cast iron and steel corrosion

\* : copperchanged color

cl : cloudy

bk : black

bw : brown

Table 17

	Example						
	97	98	99	100	101	102	103
Composition of antifreeze (% by weight)	95	95	95	95	95	95	95
Monoethylene glycol	5	5	5	5	5	5	5
Water	1.5	2.0	1.5	1.5	1.5	1.5	1.5
Phosphoric acid (85% by weight)	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Potassium phosphate II	0.02	0.02	0.02	0.02	0.005	0.05	0.02
Malonic acid	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Magnesium nitrate	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Mercaptobenzothiazole soda	4.0	4.0	4.0	4.0	4.0	4.0	4.0
Sodium nitrate	0.05	0.05	0.05	0.05	0.05	0.05	0.05
Sodium benzoate	1.8	0.8	1.8	1.8	1.8	1.8	1.0
Copolymer (1)							
Copolymer (2)							
Copolymer (3)							
Potassium hydroxide							
Sodium hydroxide							
Test for metal corrosion (mg/cm <sup>2</sup> )							
Cast aluminum	-0.02	-0.02	-0.04	-0.03	-0.06	-0.05	-0.06
Cast iron	-0.05	-0.00	-0.00	-0.07	-0.05	-0.04	-0.06
Steel	-0.06	-0.07	-0.07	-0.02	-0.02	-0.00	-0.05
Brass	-0.07	-0.03	-0.03	-0.08	-0.00	-0.03	-0.03
Solder	-0.09	-0.07	-0.09	-0.05	-0.07	-0.05	-0.09
Copper	-0.08	-0.05	-0.07	-0.02	-0.01	-0.04	-0.03
Appearance	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion
Test for corrosion of heat Transferring surface							
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.03	-0.06	-0.08	-0.06	-0.07	-0.10	-0.02
pH before test	7.8	7.8	7.8	7.8	7.8	7.8	7.8
pH after test	7.8	7.8	7.8	7.8	7.6	7.9	7.8
Appearance of liquid after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed
Appearance of test piece after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed

Table 17 (continued)

	Control						
	90	91	92	93	94	95	96
Composition of antifreeze (% by weight)							
Monethylene glycol	95	95	95	95	95	95	95
Water	5	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Potassium phosphate, II							
Malonic acid		0.5	0.5	0.5	0.5	0.5	0.5
Magnesium nitrate	0.02		0.02	0.02	0.02	0.02	0.02
Mercaptothiazole soda	0.4	0.4		0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4		0.4	0.4	0.4
Sodium benzoate	4.0	4.0	4.0	4.0		4.0	4.0
Copolymer (1)	0.05	0.05	0.05	0.05	0.05		0.05
Copolymer (2)							
Copolymer (3)							
Potassium hydroxide	1.0	1.8	1.8	1.8	1.8	1.8	
Sodium hydroxide							
Test for metal corrosion (mg/cm <sup>2</sup> )							
Cast aluminum	-0.44	-1.58	-1.02	-0.58	-0.42	-1.46	-0.47
Cast iron	-0.22	-1.58	-0.78	-1.46	-1.36	-0.49	-0.53
Steel	-0.10	-0.35	-0.38	-0.13	-0.22	-0.22	-0.35
Brass	-0.11	-0.16	-0.18	-0.06	-0.12	-0.13	-0.06
Solder	-0.15	-0.22	-0.10	-0.14	-0.36	-0.43	-0.20
Copper	-0.06	-0.18	-0.42	-0.07	-0.10	-0.11	-0.09
Appearance	Note 5	Note 1	Note 1*	Note 1	Note 2	Note 2	Note 7
Test for corrosion of heat							
Transferring surface							
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.54	-1.01	-1.92	-1.21	-0.61	-0.98	-0.48
pH before test	7.8	7.8	8.2	7.9	7.7	7.7	6.3
pH after test	9.1	9.2	8.9	8.5	8.1	9.0	7.5
Appearance of liquid after test	cl	cl	cl	cl	not changed	cl	not changed
Appearance of test piece after test	bk	bk	bk	bk	bw	bk	bw

Note 1 : cast aluminum and cast iron corrosion

Note 2 : cast aluminum, cast iron and solder corrosion

Note 5 : squeezeout corrosion of cast aluminum

Note 7 : cast iron and steel corrosion

\* : copperchanged color

cl : cloudy

bk : black

bw : brown

Table 18

	Example									
	104	105	106	107	108	109	110			
Composition of antifreeze (% by weight)	95	95	95	95	95	95	95			
Monothylene glycol	5	5	5	5	5	5	5			
Water	1.5	2.0	1.5	1.5	1.5	1.5	1.5			
Phosphoric acid (85% by weight)	0.3	0.3	0.3	0.3	0.3	0.3	0.3			
Potassium phosphate II	0.02	0.02	0.02	0.02	0.005	0.05	0.02			
Phthalic acid	0.4	0.4	0.4	0.4	0.4	0.4	0.4			
Isophthalic acid	0.4	0.4	0.4	0.4	0.4	0.4	0.4			
Magnesium nitrate	4.0	4.0	4.0	4.0	4.0	4.0	4.0			
Mercaptobenzothiazole soda	0.05	0.05	0.05	0.05	0.05	0.05	0.05			
Sodium nitrate										
Sodium benzoate										
Copolymer (1)										
Copolymer (2)										
Copolymer (3)										
Potassium hydroxide	1.8	0.8	1.8	1.8	1.8	1.8	1.8			
Sodium hydroxide										
Test for metal corrosion (mg/cm <sup>2</sup> )										
Cast aluminum	-0.09	-0.06	-0.00	-0.07	-0.01	-0.09	-0.06			
Steel	-0.00	-0.04	-0.00	-0.06	-0.06	-0.08	-0.01			
Brass	-0.08	-0.06	-0.03	-0.02	-0.05	-0.03	-0.02			
Solder	-0.09	-0.07	-0.08	-0.05	-0.05	-0.06	-0.05			
Copper	-0.06	-0.06	-0.04	-0.09	-0.07	-0.04	-0.02			
Appearance	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion			
Test for corrosion of heat transferring surface										
Change in mass of test piece (mg/cm <sup>2</sup> )	-0.02	-0.02	-0.01	-0.08	-0.01	-0.05	-0.04			
pH before test	7.8	7.8	7.8	7.8	7.8	7.8	7.8			
pH after test	7.9	7.8	7.7	7.8	7.7	7.9	7.8			
Appearance of liquid after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed			
Appearance of test piece after test	not changed	not changed	not changed	not changed	not changed	not changed	not changed			

Table 18 (continued)

	Control				
	97	98	99	100	101
Composition of antifreeze (% by weight)					
Monethylene glycol	95	95	95	95	95
Water	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5	1.5
Potassium phosphate II					
Phthalic acid		0.3	0.3	0.3	0.3
Isophthalic acid					
Magnesium nitrate	0.02		0.02	0.02	0.02
Mercaptobenzothiazole soda	0.4	0.4			
Sodium nitrate	0.4	0.4		0.4	0.4
Sodium benzoate	4.0	4.0		4.0	4.0
Copolymer (1)	0.05	0.05	0.05		0.05
Copolymer (2)					
Copolymer (3)					
Potassium hydroxide	1.0	1.8	1.8	1.8	
Sodium hydroxide					
Test for metal corrosion (mg/cm <sup>2</sup> )					
Cast aluminum	-0.34	-1.86	-1.75	-0.68	-0.42
Cast iron	-0.19	-0.79	-1.18	-0.96	-0.56
Steel	-0.10	-0.28	-0.42	-0.23	-0.33
Brass	-0.09	-0.16	-0.38	-0.08	-0.10
Solder	-0.13	-0.26	-0.45	-0.34	-0.21
Copper	-0.06	-0.14	-0.36	-0.10	-0.05
Appearance	Note 5	Note 1	Note 2*	Note 1	Note 7
Test for corrosion of heat transferring surface					
Change in mass of test piece (mg/cm <sup>2</sup> )	-1.42	-2.98	-0.92	-0.81	-0.51
pH before test	7.9	7.7	7.6	7.9	6.2
pH after test	9.0	9.3	7.9	8.2	7.6
Appearance of liquid after test	cl	cl	cl	cl	not changed
Appearance of test piece after test	bk	bk	bw	bk	bw

Note 1 : cast aluminum and cast iron corrosion

Note 2 : cast aluminum, cast iron and solder corrosion

Note 5 : squeezeout corrosion of cast aluminum

Note 7 : cast iron and steel corrosion

\* : copper changed color

cl : cloudy

bk : black

bw : brown

Examples 111 to 117

5 Varying components indicatd in Table 19 were dissolved in varying proportions indicated in Table 19 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed. The varying samples thus obtained were subjected to the following test and relevant measurement.

Test for metal corrosion

The results were as shown in Table 19.

10 Controls 102 to 105

Varying components indicated in Table 19 were dissolved in varying proportions indicated in Table 19 in 5 parts by weight of tap water or 95 parts by weight of monoethylene glycol and the two liquids were mixed.

The varying samples thus obtained were subjected to the following test and relevant measurement.

15 Test for metal corrosion

The results were as shown in Table 19.

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Table 19

	Example						
	111	112	113	114	115	116	117
Composition of antifreeze (% by weight)							
Moethylene glycol	95	95	95	95	95	95	95
Water	5	5	5	5	5	5	5
Phosphoric acid (85% by weight)	1.5		1.5	1.5	1.5	1.5	1.5
Potassium phosphate II		2.0					
Magnesium nitrate	0.02	0.02	0.02	0.02	0.005	0.05	0.02
Macrcaptobenzothiazole soda	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Sodium nitrate	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Sodium benzoate	4.0	4.0	4.0	4.0	4.0	4.0	4.0
Copolymer (1)	0.05	0.05			0.05	0.05	0.05
Copolymer (2)							
Copolymer (3)							
Potassium hydroxide	1.0	0.4	1.0	1.0	1.0	1.0	
Sodium hydroxide							
Test for metal corrosion (mg/cm <sup>2</sup> )							
Cast aluminum	-0.06	-0.03	-0.00	-0.03	-0.02	-0.07	-0.04
Cast aluminum	-0.00	-0.02	-0.07	-0.03	-0.01	-0.05	-0.06
Steel	-0.04	-0.02	-0.05	-0.04	-0.05	-0.02	-0.01
Brass	-0.07	-0.04	-0.05	-0.04	-0.05	-0.06	-0.05
Solder	-0.07	-0.10	-0.08	-0.10	-0.02	-0.10	-0.03
Copper	-0.04	-0.02	-0.03	-0.03	-0.06	-0.02	-0.03
Appearance of test piece	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion	no corrosion

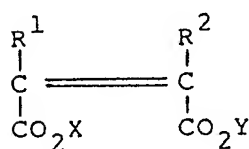
Table 19 (continued)

	Control			
	102	103	104	105
Composition of antifreeze (% by weight)				
Moethylene glycol Water	95	95	95	95
Water	5	5	5	5
Phosphoric acid (85% by weight)	1.5	1.5	1.5	1.5
Potassium phosphate II				
Magnesium nitrate		0.02	0.02	0.02
Macrcaptobenzothiazole soda	0.4		0.4	0.4
Sodium nitrate	0.4		0.4	0.4
Sodium benzoate	4.0		4.0	4.0
Copolymer (1)	0.05	0.05		0.5
Copolymer (2)				
Copolymer (3)				
Potassium hydroxide	1.0	1.0	1.0	
Sodium hydroxide				
Test for metal corrosion (mg/cm <sup>2</sup> )				
Cast aluminum	-1.28	-1.02	-0.68	-0.32
Cast aluminum	-0.12	-1.40	-1.21	-0.61
Steel	-0.52	-0.56	-0.26	-0.23
Brass	-0.09	-0.19	-0.10	-0.08
Solder	-0.22	-0.42	-0.27	-0.21
Copper	-0.11	-0.34	-0.10	-0.10
Appearance of test piece	corrosion	corrosion	corrosion	corrosion



## Claims

- 5
1. An antifreeze composition comprising a glycol and water, characterized by containing
- (A) a phosphoric acid compound,
- (B) a compound of at least one metal selected from the group consisting of manganese and magnesium, and 10
- (C) a corrosionproofing agent, and having a pH value in the range of 6.5 to 9.
2. A composition according to Claim 1, characterized in that the gravimetric ratio of said glycol to water is in the range of 99 : 1 to 5 : 95.
3. A composition according to Claim 2, characterized in that (A) said phosphoric acid compound is contained in a proportion in the range of 0,1 to 5% by weight, (B) said manganese compound in a proportion of 0,0001 to 0,05% by weight or said magnesium compound in a proportion in the range of 0,001 to 0,08% by weight, and (C) said corrosionproofing agent in a proportion in the range 0,05 to 5% by weight, respectively based on the amount of the glycol. 15
4. A composition according to Claim 3, characterized in that (C) said corrosionproofing agent comprises sodium mercaptothiazole, a nitrate, and a benzoate in respective proportions in the range of 0,05 to 0,7% by weight, 0,05 to 0,7% by weight, and 1 to 7% by weight, based on the glycol. 20
5. An antifreeze composition comprising a glycol and water, characterized by containing
- (A) a phosphoric acid compound,
- (B) a compound of at least one metal selected from the group consisting of manganese and magnesium, 25
- (C) a corrosionproofing agent, and
- (D) at least one compound selected from the group consisting of
- (a) an aromatic polybasic acid or a salt thereof,
- (b) an aliphatic dicarboxylic acid or a salt thereof,
- (c) a molybdate, 30
- (d) a tungstate,
- (e) a homopolymer derived from an alkylene glycol monoallyl ether (I) represented by the formula
- I:
- $$\text{CH}_2 = \text{CH}-\text{CH}_2\text{O}(\text{C}_2\text{H}_4\text{O})_m(\text{C}_3\text{H}_6\text{O})_n\text{H} \quad (\text{I})$$
- wherein m et n are independently 0 or a positive integer, providing that the sum of m and n falls in the range of 1 to 100 and the (C<sub>2</sub>H<sub>4</sub>O) units and the (C<sub>3</sub>H<sub>6</sub>O) units may be bound in any order, or a maleic acid type monomer (II) represented by the formula II:
- 35



(II)

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wherein R<sup>1</sup> and R<sup>2</sup> are independently hydrogen atom or methyl group, and X and Y are independently (C<sub>2</sub>H<sub>4</sub>O)<sub>p</sub>(C<sub>3</sub>H<sub>6</sub>O)<sub>q</sub>R<sup>3</sup>, wherein R<sup>3</sup> is alkyl group of 1 to 20 carbon atoms and p and q are independently 0 or a positive integer, providing that the sum of p and q falls in the range of 0 to 100 and the (C<sub>2</sub>H<sub>4</sub>O) units and the (C<sub>3</sub>H<sub>6</sub>O) units may be bound in any order, monovalent metal, divalent metal, ammonium group, or organic amine group, and 50

- (f) a copolymer derived from the alkylene glycol monoallyl ether (I) or the maleic acid type monomer (II) with a monomer (III) copolymerizable therewith, having a pH value in the range of 6.5 to 9.
6. A composition according to Claim 5, characterized in that the gravimetric ratio of said glycol to water is in the range of 99 : 1 to 5 : 95. 55
7. A composition according to Claim 6, characterized in that (A) said phosphoric acid compound is contained in a proportion in the range of 0.1 to 5% by weight, (B) said manganese compound in a proportion of 0,0001 to 0,05% by weight or said magnesium compound in a proportion in the range of 0,001 to 0,08% by weight, and (C) said corrosionproofing agent in a proportion in the range 0,05 to 5% by weight, respectively based on the amount of the glycol. 60
8. A composition according to Claim 7, characterized in that (C) said compound is an aromatic polybasic acid and is incorporated in a proportion in the range of 0,05 to 1% by weight, based on the amount of the glycol.
9. A composition according to Claim 7, characterized in that (C) said compound is an aliphatic 65

dicarboxylic acid and is incorporated in a proportion in the range of 0,05 to 1% by weight, based on the amount of the glycol.

10. A composition according to Claim 7, characterized in that (C) said compound is a molybdate and is incorporated in a proportion in the range of 0,1 to 1% by weight, based on the amount of the glycol.

11. A composition according to Claim 7, characterized in that (C) said compound is a tungstate and is incorporated in a proportion in the range of 0,1 to 1 % by weight, based on the amount of the glycol.

12. A composition according to Claim 7, characterized in that (C) said compound is a homopolymer of at least one monomer selected from the group consisting of alkylene glycol monoallyl ethers (I) and maleic acid type monomers (II) or a copolymer of said monomer with a monomer (III) copolymerizable therewith and is incorporated in a proportion of not less than 0,01% by weight based on the amount of the glycol.

13. A composition according to Claim 7, characterized in that said corrosionproofing agent is at least one compound selected from the group consisting of sodium mercaptobenzothiazole, methyl benzotriazole, sodium nitrite, and sodium benzoate.